

# Uncovering the source of the anomaly in the electrical resistivity of ferromagnetic metals around liquid-helium temperatures

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Ferromagnetism is an exciting phase of matter exhibiting strongly correlated electron behavior and a standard example of spontaneously broken rotational symmetry. Below the Curie temperature, atomic magnets in ferromagnetic metals align along a specific direction and the scattering of conduction electrons from thermal perturbations to this spin order, together with electron-electron collisions, mark the material electrical behavior at low temperatures. However, around liquid-helium temperatures, a departure from this well-established picture exists, observed as a *linear* variation of the electrical resistivity with temperature, which has puzzled scientists for a long time. Here we introduce a spin-flip scattering mechanism of conduction electrons in ferromagnetic metals arising from their interaction with the internal magnetic induction and mediated by chiral modes of the crystal lattice vibrations carrying spin 1. This mechanism is able to explain the above anomaly and give a good account of the spin-lattice relaxation times of iron, cobalt and nickel at room temperatures.

## I. INTRODUCTION

For decades, the microscopic process which causes a linear-in-temperature term in the electrical resistivity of ferromagnetic metals at low temperatures, which is clearly observed around liquid-helium temperatures,<sup>1,2</sup> has remained unclear. In this temperature region, the  $T^2$  dependence of the electrical resistivity characteristic of the transition metals at low temperatures, due to the  $s$ - $d$  exchange interaction<sup>3-5</sup> and inter-electronic collisions,<sup>6</sup> ceases to be the only dominant contribution. The most known intrinsic mechanism giving a linear term in the resistivity is the spin-orbit interaction between the orbits of the  $4s$  conduction electrons and the spins of the nearly localized  $3d$  ferromagnetic electrons.<sup>7-9</sup> However, this predicts a linear coefficient which is about a thousand of times smaller than observed.<sup>4,8,10</sup>

Despite other mechanisms have been proposed<sup>2</sup> to explain this anomalous behavior, including e.g. electron-magnon scattering taking into account the electronic spin polarization, and scattering of the conduction electrons by 2D spin-wave excitations on the magnetic domain walls; it is believed,<sup>1,2</sup> based on a series of experiments, that the anomaly is caused by the scattering of conduction electrons by the internal magnetic induction present in the ferromagnetic metals, observed as an internal magnetoresistance effect. However, no explanation of this fact has been given so far using quantum mechanics.

In this article, we introduce a simple picture of an internal magnetoresistance effect in the ferromagnetic metals giving the correct magnitude of the linear coefficient. This consists of electronic spin-flip transitions within the Zeeman energy produced by the internal magnetic induction and mediated by the isotropic (spin-phonon) interaction of the conduction electrons with the contact field they produce at the lattice sites of the crystal while they are in orbital motion with the atoms. This mechanism, which accounts for the observed spin-lattice relaxation times of pure ferromagnetic metals at room tem-

peratures, complements the existing theories of spin relaxation of conduction electrons in metals,<sup>11-14</sup> which do not deal with the ferromagnetic case.

Our analysis begins by noting that, as observed e.g. from de Haas-van Alphen oscillation experiments,<sup>15-17</sup> the internal magnetic induction seen by conduction electrons in the ferromagnetic metals within each magnetic domain, and even in the absence of an external magnetic field as we consider here, is known to be

$$\mathbf{B}_i = 4\pi \mathbf{M}_s, \quad (1)$$

where  $\mathbf{M}_s$  is the magnetization within each magnetic domain, which is typically measured to be the saturation magnetization.<sup>18</sup> We imagine the unmagnetized material as a collection of magnetic domains and, since electronic scattering off the domain walls do not appreciably contribute to the observed anomaly, we concentrate only on one of the domains, where a quantization direction is given by that parallel to the majority spins, i.e. the spin up direction,  $-\mathbf{M}_s/M_s$ , for the electrons assuming spin-orbit coupling is negligible.

Now the internal magnetic induction arises from the magnetic fields associated with the magnetic dipole moments of the ferromagnetic atoms, whereas the alignment of these moments within each magnetic domain is overwhelmingly electrostatic in nature, due to electron-electron exchange interactions, whose characteristic energies are thousands of times greater than the dipolar energies. So how can the internal magnetic induction in ferromagnetic metals be the source of an anomaly in an environment for the conduction electrons dominated by much greater effective exchange fields?

To address this question we consider the electrical conduction by a ferromagnetic metal, assuming that the current is entirely transported by the  $4s$  electrons. The kinetic and exchange energies of these electrons are described here by the mean-field Hamiltonian<sup>19,20</sup>

$$\hat{H}_{\text{kin+ex}} = \sum_{\mathbf{k}s} \varepsilon_{\mathbf{k}s}^0 \hat{c}_{\mathbf{k}s}^\dagger \hat{c}_{\mathbf{k}s}, \quad \varepsilon_{\mathbf{k}s}^0 = \frac{\hbar^2 k^2}{2m} - s(\Delta_{\text{ex}}/2), \quad (2)$$

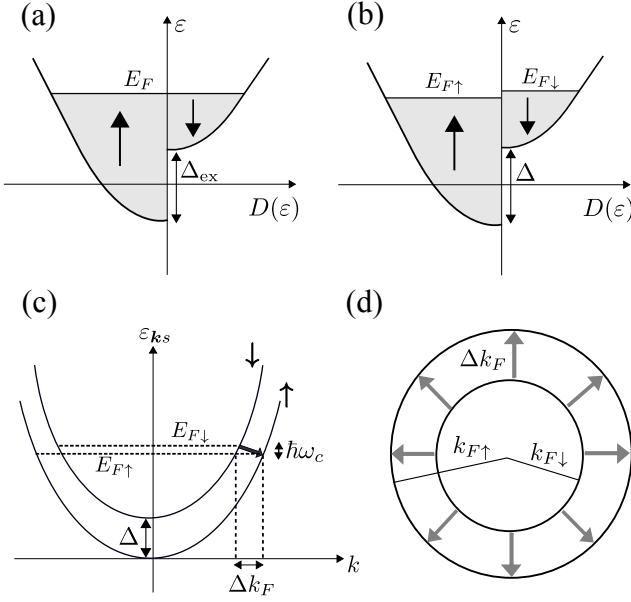


FIG. 1. Electron spin relaxation in the presence of an internal magnetic induction. (a) Electronic density of states in the presence of exchange interactions and no internal magnetic induction. (b) The internal induction is suddenly turned on, with  $\Delta = \Delta_{\text{ex}} + \hbar\omega_c$ , creating an initial Fermi surface instability  $E_{F\downarrow} - E_{F\uparrow} = \hbar\omega_c$ . (c) Spin-split bands showing the most probable processes restoring the initial equilibrium. (d) Fermi surfaces of the two spin populations in  $k$ -space, showing the minimum crystal momentum transfer  $\Delta k_F$ . The Fermi surfaces are drawn with a spherical shape for simplicity, but this assumption is relaxed in the main text.

where  $\hat{c}_{\mathbf{k}s}^\dagger$  ( $\hat{c}_{\mathbf{k}s}$ ) is the creation (annihilation) operator for a conduction electron with wavevector  $\mathbf{k}$  and spin index  $s$ ,  $\Delta_{\text{ex}}$  is the exchange spin-splitting of the conduction band,  $m$  is the electron effective mass and, when not a subindex,  $s = \pm$  according to a conduction electron having its spin  $\uparrow$  or  $\downarrow$  with respect to the quantization direction.

Fig 1(a) shows a cartoon of the conduction electron spin polarization produced by the electron-electron exchange interactions in the absence of internal magnetic induction. Now if this magnetic induction is turned on, no matter how small it is compared to the exchange fields, a slight Zeeman shift of the original energy bands

$$\varepsilon_{\mathbf{k}s} = \varepsilon_{\mathbf{k}s}^0 - s(\hbar\omega_c/2), \quad \hbar\omega_c = 2\mu_B B_i, \quad (3)$$

is evidently produced, where  $\omega_c$  is the conduction electron cyclotron frequency due to the internal magnetic induction  $B_i = 4\pi M_s$  in (1), with  $\mu_B$  being the Bohr magneton. This disturbs the equilibrium of the spin populations imposed by the exchange interactions, as shown schematically in Fig 1(b).

The conduction electron spin relaxation processes that we describe here is a way for the electronic system to restore the initial equilibrium imposed by the exchange fields, with the most probable spin-flip transitions at low

enough temperatures involving an energy loss  $\hbar\omega_c$  and the minimum allowed crystal momentum transfer  $\Delta k_F$  as shown in Figs. 1(c) and 1(d), and demonstrated later. Clearly, these require the mediation of a type of elementary excitations, other than spin waves in the 3d ferromagnetic electrons, capable of absorbing the change of energy, linear and spin angular momentum of the electronic system for each spin-flip scattering event.

## II. CONTACT FIELD FROM ELECTRON-ATOM COMOTION

A clue for the appearance of such excitations arises when the exercise of exhausting all sources of dipolar fields originating from the conduction electrons is done. Among these, the magnetic effects of the contact field at the crystal lattice sites

$$\hat{B}_c(\mathbf{x}) = -\frac{8\pi}{3}\mu_B \sum_n \delta(\mathbf{x} - \mathbf{R}_n) \hat{\mathbf{L}}_n, \quad (4)$$

created from the *orbital* comotion of the conduction electrons with the atoms, have been overlooked in the theories of spin relaxation of conduction electrons in metals but shall be proved here to play a dominant role for the mediation of the transitions in Fig 1.

The operator  $\hat{\mathbf{L}}_n = \hat{\mathbf{r}}_n \times \hat{\mathbf{p}}_n$  in (4) is the orbital angular momentum (in units of  $\hbar$ ) of the  $n^{\text{th}}$  atom, and hence of a conduction electron *rigidly* comoving with it for a long enough time, where  $\hat{\mathbf{r}}_n$  is therefore the displacement of the atom from the lattice point  $\mathbf{R}_n$ , and  $\hat{\mathbf{p}}_n = M\dot{\hat{\mathbf{r}}}_n$ , with  $M$  the atomic mass. The negligible relative motion of the conduction electrons with respect to the atom it temporarily “belongs to” is suggested by the neglect of the spin-orbit interaction being a good approximation in ferromagnetic metals<sup>21</sup> and implies that, after quantizing the atomic orbits,<sup>22</sup> the electron orbital angular momentum  $\hat{\mathbf{L}}_n = (1/N) \sum_{\mathbf{q}\mathbf{q}'} \hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}'} e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{R}_n}$ , can be expressed (see Appendix B) in terms of

$$\hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}'} = \frac{1}{2} \sum_{\alpha\alpha'} \sqrt{\frac{\omega_{\mathbf{q}'\alpha'}}{\omega_{\mathbf{q}\alpha}}} (\hat{a}_{\mathbf{q}\alpha} + \hat{a}_{-\mathbf{q}\alpha}^\dagger) \mathbf{S}_{\mathbf{q}\mathbf{q}'(\alpha\alpha')} \times (\hat{a}_{-\mathbf{q}'\alpha'} - \hat{a}_{\mathbf{q}'\alpha'}^\dagger), \quad (5)$$

where  $N$  is the number of atoms,  $\hat{a}_{\mathbf{q}\alpha}^\dagger$  ( $\hat{a}_{\mathbf{q}\alpha}$ ) is the creation (annihilation) operator of a phonon with wavevector  $\mathbf{q}$ , angular frequency  $\omega_{\mathbf{q}\alpha}$ , and *linear* polarization in the direction of the real unit vector  $\mathbf{e}_{\mathbf{q}\alpha}$ , and  $\mathbf{S}_{\mathbf{q}\mathbf{q}'(\alpha\alpha')} = -i(\hat{\mathbf{e}}_{\mathbf{q}\alpha} \times \hat{\mathbf{e}}_{\mathbf{q}'\alpha'})$ . Note the independence of (5) on the atomic mass, making plausible referring to  $\hat{\mathbf{L}}_n$  as the orbital angular momentum of a conduction electron comoving with the  $n^{\text{th}}$  atom. For the phonon dispersion, we take the isotropic Debye spectrum with transverse  $\omega_{\mathbf{q}1,2} = \omega_{\mathbf{q}T} = c_T q$  and longitudinal  $\omega_{\mathbf{q}3} = \omega_{\mathbf{q}L} = c_L q$  branches.

We remark that the possibility of conduction electrons collectively “belonging to” different atoms for long

enough times, despite their band motion, may arise due to the strongly correlated electron behavior induced by the electron-electron exchange interactions<sup>23</sup> and, as pointed out by Zener<sup>24</sup> long ago, the fact that the 4s conduction electrons spend a greater fraction of their time within the region of the 3d shells (which are somewhat “localized” to the atoms) when they are in the solid than when they are in an isolated atom. We shall show later after (13) that a resonance condition where the atoms rotate in a transverse phonon mode with the electron cyclotron frequency is possible around liquid-helium temperatures, supporting the above picture, which may be thought of as a modified Drude picture of conduction electrons colliding with atoms while they conduct the electricity, but with a *finite* duration of a collision during which the electron and atom share their orbital motion; this duration can be estimated to be about a tenth or less of the Drude relaxation time for the ferromagnetic elements at low temperatures (see Appendix B)

### III. SPIN OF THE PHONON

With the notation  $\mathbf{S}_{\mathbf{q}(\alpha\alpha')} = \mathbf{S}_{\mathbf{q}\mathbf{q}(\alpha\alpha')}$ , we can write the total angular momentum of the phonon system as  $\hat{\mathbf{S}}_{\text{ph}} \equiv \sum_n \hat{\mathbf{L}}_n = \sum_{\mathbf{q}} \hat{\mathbf{S}}_{\mathbf{q},\text{ph}}$ , where the angular momentum operator of a single phonon,  $\hat{\mathbf{S}}_{\mathbf{q},\text{ph}} = \hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}}$ , with wave vector  $\mathbf{q}$  is given by

$$\hat{\mathbf{S}}_{\mathbf{q},\text{ph}} = \frac{1}{2} \sum_{\alpha\alpha'} \sqrt{\frac{\omega_{\mathbf{q}\alpha'}}{\omega_{\mathbf{q}\alpha}}} (\hat{a}_{\mathbf{q}\alpha} + \hat{a}_{-\mathbf{q}\alpha}^\dagger) \mathbf{S}_{\mathbf{q}(\alpha\alpha')} (\hat{a}_{-\mathbf{q}\alpha'} - \hat{a}_{\mathbf{q}\alpha'}^\dagger). \quad (6)$$

The sum in this expression can be seen as a matrix multiplication. In fact, with  $\mathbf{e}_{q1} = (1, 0, 0)$ ,  $\mathbf{e}_{q2} = (0, 1, 0)$ , and  $\mathbf{e}_{q3} = \mathbf{q}/q = (0, 0, 1)$ , the matrices  $\mathbf{S}_{\mathbf{q}}$  constitute a representation of the infinitesimal generators of rotations in three dimensions, with  $\mathbf{S}_{\mathbf{q}}^2 = \sum_a \mathbf{S}_{\mathbf{q}}^a \mathbf{S}_{\mathbf{q}}^a = 2 \cdot \mathbf{1}$ , and the commutation relations  $[\mathbf{S}_{\mathbf{q}}^a, \mathbf{S}_{\mathbf{q}}^b] = i \sum_c \epsilon_{abc} \mathbf{S}_{\mathbf{q}}^c$ , and  $[\mathbf{S}_{\mathbf{q}}^a, \mathbf{S}_{\mathbf{q}}^a] = 0$ .

As usual in the theory of angular momentum, it is convenient to work in a representation which simultaneously diagonalizes  $\mathbf{S}_{\mathbf{q}}^3$  and  $\mathbf{S}_{\mathbf{q}}^2$ . This is done by changing basis from  $\{\mathbf{e}_{\mathbf{q}\alpha}\}$  to the helicity basis  $\{\mathbf{e}_{\mathbf{q}\sigma}\}$  defined by the circular  $\mathbf{e}_{\mathbf{q}\pm} = (1/\sqrt{2})(\mathbf{e}_{q2} \mp i\mathbf{e}_{q1})$  and longitudinal  $\mathbf{e}_{q0} = i\mathbf{e}_{q3}$  polarization vectors, where  $\mathbf{e}_{-\mathbf{q}\sigma}^* = \mathbf{e}_{\mathbf{q}\sigma}$ . The corresponding unitary transformation can be shown to map the matrices  $\mathbf{S}_{\mathbf{q}(\alpha\alpha')}$  to  $\mathbf{S}_{\mathbf{q}(\sigma\sigma')} = -i(\hat{\mathbf{e}}_{\mathbf{q}\sigma}^* \times \hat{\mathbf{e}}_{\mathbf{q}\sigma'})$ , which are the spin matrices for a spin-1 particle (see Appendix B). We have here the analog of circularly polarized photons in vacuum, with the difference that in the phonon system longitudinal excitations are possible.

In the new representation, the *chiral* phonon operators are  $\hat{b}_{\mathbf{q}\pm} = (1/\sqrt{2})(\hat{a}_{q2} \mp i\hat{a}_{q1})$ , which annihilate phonons with  $\pm$  helicities (circular polarization); and  $\hat{b}_{q0} = i\hat{a}_{q3}$ , which annihilates zero helicity (or longitudinal) modes. We have, for instance,  $\hat{\mathbf{S}}_{\mathbf{q},\text{ph}}^3 = \hat{b}_{\mathbf{q}+}^\dagger \hat{b}_{\mathbf{q}+} - \hat{b}_{\mathbf{q}-}^\dagger \hat{b}_{\mathbf{q}-}$ . These phonons have definite spin projections  $\pm 1, 0$  along the

propagation direction and, making an analogy with circularly polarized light, the displacement field of the atoms must rotate perpendicular to the propagation direction in a circularly polarized elastic wave.<sup>25</sup> We use the convention  $\mathcal{PT} \hat{b}_{\mathbf{q}\sigma} = \hat{b}_{-\mathbf{q}\sigma}^* = \hat{b}_{\mathbf{q}\sigma}$ , inherited from  $\mathbf{e}_{-\mathbf{q}\sigma}^* = \mathbf{e}_{\mathbf{q}\sigma}$ , which states that the wavefunctions of the crystal lattice vibrations are even under the  $\mathcal{PT}$  transformation (complex conjugation + parity).

When the transformation to the helicity basis is performed in the tensor product spin space corresponding to  $\mathbf{q}$  and  $\mathbf{q}'$ , we take  $\mathbf{S}_{\mathbf{q}\mathbf{q}'} \rightarrow \mathbf{S}_{\mathbf{q}} \otimes \mathbf{1}_{\mathbf{q}'} = \mathbf{1}_{\mathbf{q}} \otimes \mathbf{S}_{\mathbf{q}'}$  in (5), since the image of  $\mathbf{S}_{\mathbf{q}\mathbf{q}'}$  under the transformation must behave as an angular momentum upon rotations and, by definition, must reduce to  $\mathbf{S}_{\mathbf{q}}$  when  $\mathbf{q} = \mathbf{q}'$ . In the helicity basis, we then express the ladder operators corresponding to (5) as

$$\hat{S}_{\mathbf{q}\mathbf{q}'}^- = \frac{1}{2} \left[ \sqrt{\frac{2\omega_{\mathbf{q}'L}}{\omega_{\mathbf{q}T}}} (\hat{b}_{\mathbf{q}-}^\dagger + \hat{b}_{\mathbf{q}+}) (\hat{b}_{\mathbf{q}'0}^\dagger + \hat{b}_{\mathbf{q}'0}) - \sqrt{\frac{2\omega_{\mathbf{q}'T}}{\omega_{\mathbf{q}L}}} (\hat{b}_{\mathbf{q}0}^\dagger - \hat{b}_{\mathbf{q}0}) (\hat{b}_{\mathbf{q}'-}^\dagger - \hat{b}_{\mathbf{q}'-}) \right], \quad (7)$$

and  $\hat{S}_{\mathbf{q}\mathbf{q}'}^+ = (\hat{S}_{\mathbf{q}\mathbf{q}'}^-)^\dagger$  which, as we will see next, enter the terms in the Hamiltonian causing spin-flip scattering events of the conduction electrons giving the desired contribution to the electrical resistivity.

Denoting the Bloch wavefunctions of the conduction electrons as  $\varphi_{\mathbf{k}s}(\mathbf{x})$ , we can express their magnetic moment density in second quantized operator notation as  $\hat{\boldsymbol{\mu}}(\mathbf{x}) = -2\mu_B \sum_{ss', \mathbf{k}\mathbf{k}'} \varphi_{\mathbf{k}'s'}^*(\mathbf{x}) \varphi_{\mathbf{k}s}(\mathbf{x}) \hat{c}_{\mathbf{k}'s'}^\dagger \boldsymbol{\sigma}_{s's} \hat{c}_{\mathbf{k}s}$  where  $\boldsymbol{\sigma}_{s's}$  are the Pauli spin-1/2 matrices. The magnetic interaction of the conduction electrons with the contact field they create at the crystal lattice sites is given by  $\hat{H}_c = -\int d\mathbf{x} \hat{\boldsymbol{\mu}}(\mathbf{x}) \cdot \hat{\mathbf{B}}_c(\mathbf{x})$ , and we are particularly interested in those terms able to mediate the spin-flip transitions of Fig. 1. By using the Bloch theorem for  $\varphi_{\mathbf{k}s}(\mathbf{x}) = e^{i\mathbf{k}\cdot\mathbf{x}} u_{\mathbf{k}s}(\mathbf{x})$ , i.e.  $u_{\mathbf{k}s}(\mathbf{x} + \mathbf{R}_n) = u_{\mathbf{k}s}(\mathbf{x})$  to express  $u_{\mathbf{k}s}(\mathbf{R}_n) = u_{\mathbf{k}s}(0) = \varphi_{\mathbf{k}s}(0)$ , and neglecting umklapp processes, we have for these terms mainly

$$\hat{H}_c^{\dagger\dagger} = \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}', \mathbf{q}\mathbf{q}'} A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow} \delta_{\Delta\mathbf{q}, -\Delta\mathbf{k}} \hat{c}_{\mathbf{k}'\uparrow}^\dagger \hat{c}_{\mathbf{k}\downarrow} \hat{S}_{\mathbf{q}\mathbf{q}'}^-, \quad (8)$$

where  $\Delta\mathbf{k} = \mathbf{k}' - \mathbf{k}$ ,  $\Delta\mathbf{q} = \mathbf{q}' - \mathbf{q}$ , and we have defined the matrix elements  $A_{\mathbf{k}s, \mathbf{k}'s'} = -\frac{16\pi}{3} \mu_B^2 \varphi_{\mathbf{k}'s'}^*(0) \varphi_{\mathbf{k}s}(0)$  giving the strength of the resulting hyperfine coupling. Eq. (8) accounts for processes where spin- $\downarrow$  electrons “decay” to the majority-spin band. The reverse processes can be shown to be exponentially suppressed at low temperatures (see Appendix C).

A calculation of the spin-lattice relaxation time can then be carried out using standard perturbation theory by imagining the electrons and phonons uncoupled for times  $t < 0$  in a local equilibrium<sup>26</sup> with Hamiltonian  $\hat{\mathcal{H}}_0$

$$\hat{\mathcal{H}}_0 = \hat{H}_{\text{kin+ex}} + \sum_{\mathbf{q}\sigma} \hbar\omega_{\mathbf{q}\sigma} \hat{b}_{\mathbf{q}\sigma}^\dagger \hat{b}_{\mathbf{q}\sigma}, \quad (t < 0), \quad (9)$$

$$\hat{H} = \hat{\mathcal{H}}_0 + \hat{H}_1 + \hat{H}_c, \quad (t \geq 0). \quad (10)$$

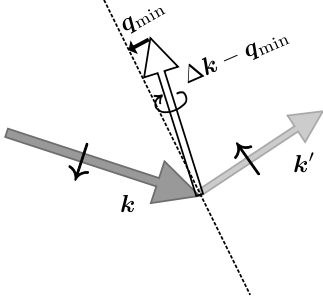


FIG. 2. Sketch of the most relevant kind of transitions (in  $k$ -space) involved in the conduction electron spin relaxation at very low temperatures. The dashed line is in the direction of  $\Delta \mathbf{k} = \mathbf{k}' - \mathbf{k}$  and  $\mathbf{q}_{\min}$  is the wavevector corresponding to the lowest energy phonon mode supported by the crystal lattice. Small arrows superposed on big arrows indicate spin states.

At times  $t \geq 0$  the internal magnetic induction is turned on as well as the contact interaction. The former magnetically couples as  $\hat{H}_i = -\int d\mathbf{x} \hat{\boldsymbol{\mu}}(\mathbf{x}) \cdot \mathbf{B}_i(\mathbf{x})$  with the conduction electron spins and has the effect of shifting the electronic energy bands from (2) to (3) giving rise to the initial Fermi surface instability: electrons remain in equilibrium with those of the same spin species but in a nonequilibrium state with those of the opposite spin. The magnetic interaction of the conduction electrons with the contact field they create at the lattice sites (these being the equilibrium positions of the atomic nuclei) is treated as the perturbation restoring the initial equilibrium imposed by the exchange fields, mainly by means of terms like (8) bringing occupied spin- $\downarrow$  electrons to unoccupied spin- $\uparrow$  states.

We remark that no further shift to the electronic energy bands is caused by the contact interaction since its diagonal matrix elements in the space of the electrons  $s \mu_B A_{\mathbf{k}s, \mathbf{k}s} \bar{S}_{\text{ph}}^3$  vanish, where  $\bar{S}_{\text{ph}}^3 = \bar{N}_+ - \bar{N}_-$  is the thermal-averaged total phonon angular momentum along the quantization direction, expressed (in units of  $\hbar$ ) in terms of the thermal-averaged number of circular phonons  $\bar{N}_{\pm} = \sum_{\mathbf{q}} \bar{N}_{\mathbf{q}\pm}$  at temperature  $T$ , with  $\bar{N}_{\mathbf{q}\pm}$  the Bose distribution and  $\bar{N}_{\mathbf{q}\sigma}^{-1} = \exp[(\hbar\omega_{\mathbf{q}\sigma} + \Sigma_{\mathbf{q}\sigma})/k_B T] - 1$ . The vanishing is because the degeneracy of the transverse bands in the phonon spectrum in our isotropic model can only be lifted if there is an *external* magnetic field.<sup>27</sup> In its absence,  $\Sigma_{\mathbf{q}\sigma}$  is therefore zero, implying a vanishing projection of the angular momentum,<sup>22</sup>  $\bar{S}_{\text{ph}}^3 = 0$ .

#### IV. ELECTRON SPIN RELAXATION

The transition probability rate for a conduction electron undergoing a collision from the state  $\mathbf{k}\downarrow$  to an unoccupied state  $\mathbf{k}'\uparrow$  is obtained from (7) and (8), in the leading order of perturbation theory, using Fermi's golden rule.<sup>26</sup> Since for temperatures  $T \rightarrow 0$  the average occupation of a phonon mode,  $\bar{N}_{\mathbf{q}\sigma}$ , is exponentially small *except* for the lowest energy mode supported by the crystal lat-

tice, which has wavevector magnitude  $q_{\min} = 2\pi/L = 0^+$ , where  $L$  is the largest linear size of the sample (which grows without bounds in the thermodynamic limit), the processes happening more frequently at very low temperatures are those in which these lowest energy modes are involved, for which conservation of energy reads

$$\hbar c_T |\Delta \mathbf{k}| - |\Delta \epsilon_{\mathbf{k}}| = 0, \quad (11)$$

up to a negligible term of  $O(1/L)$ . Therefore, neglecting contributions from exponentially small terms as well as the temperature-independent term which contribute to the residual resistivity (discussed in Appendix C), the transition probability rate, per electron, for temperatures  $T \rightarrow 0$  is

$$w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow} = \frac{3\pi}{16} \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2 |\Delta \epsilon_{\mathbf{k}}|^2}{(k_B \Theta)^4} \frac{k_F}{|\Delta \mathbf{k}|} \frac{c_s^4}{c_L^2 c_T^2} \left(\frac{2}{z}\right)^{4/3} \times \frac{(k_B T)}{\hbar} \ln \left( \frac{k_B T}{\hbar c_L q_{\min}} \right) + O(T^2), \quad (12)$$

where  $\Theta$  is the Debye temperature,  $k_F$  is the Fermi wavevector,  $z = N_e/N$  is the number of conduction electrons per atom and  $c_s$  is average speed of sound, determined from  $3/c_s^3 = 2/c_T^3 + 1/c_L^3$ .

The dominant (first) term in (12) comes from processes wherein a lowest energy longitudinal mode is absorbed and a circular mode is *spontaneously* emitted to satisfy the conservation laws, as shown in Fig. 2. To restore the equilibrium  $E_{F\uparrow} = E_{F\downarrow} = E_F$  imposed by the exchange fields, electrons will then try to minimize energy by transferring from the minority-spin to the majority-spin band, with the most probable transition per unit time obtained, from (12), when  $|\Delta \epsilon_{\mathbf{k}}|$  is maximum and  $|\Delta \mathbf{k}|$  is minimum, i.e. when  $|\Delta \epsilon_{\mathbf{k}}| = |E_{F\uparrow} - E_{F\downarrow}| = \hbar \omega_c$  and  $|\Delta \mathbf{k}| = \Delta k_F$ , as shown in Fig. 1.

The most probable processes restoring the initial equilibrium satisfy (11) in the form  $\hbar \omega_c = \hbar c_T \Delta k_F$  or equivalently, by defining  $T_i = \hbar \omega_c / k_B$  as the characteristic temperature associated with the internal magnetic induction and  $T_{\text{res}} = \hbar c_T \Delta k_F / k_B$  as that associated with the excitation of the circular phonon modes mainly in charge of the conduction electron spin relaxation process, in the form  $T_i = T_{\text{res}}$ . By writing<sup>28</sup>  $\Delta k_F / k_F = \Delta_{\text{ex}} / 2E_F$  we can express  $T_{\text{res}}$  as

$$T_{\text{res}} = \frac{c_T}{c_s} (z/2)^{1/3} (\Delta_{\text{ex}} / 2E_F) \Theta. \quad (13)$$

As shown in table I the agreement of  $T_i = T_{\text{res}}$  is remarkably good, given our estimate of  $\Delta_{\text{ex}}$ , with the characteristic temperatures being around liquid-helium temperatures. In this regime, a *resonant* chiral mode of the lattice vibrations should then exist wherein the atoms rotate at the conduction electron cyclotron frequency due to the internal magnetic induction, which is in favor of the picture of a collective motion of electrons comoving with the atoms for sufficiently long times.

We can account for less probable transitions involving the magnitudes of energy loss and crystal momentum transfer  $0 \leq |\Delta \epsilon_{\mathbf{k}}| < \hbar \omega_c$  and  $\Delta k_F < |\Delta \mathbf{k}| \leq 2k_F$ ,



Metal	$z$	$n$ ( $10^{28}\text{m}^{-3}$ )	$c_L$ (m/s)	$T_{\text{res}}$ (°K)	$T_i$ (°K)	$\Theta$ (°K)	$\xi$	$H_a$ (MG)	$\delta\Omega_F$	$\gamma$ ( $10^{-12}\Omega\text{ cm}/^\circ\text{K}$ )		$\tau_{\downarrow\uparrow}$ (ns)	
			$c_T = c_L/\sqrt{2}$							theory	exp.	theory	exp.
Fe	0.22	8.5	5960	4.0	2.9	470	0.35	1.95	0.5	16.8	11–49.3	0.04	0.03–0.1
Co	0.7	9.1	4720	5.4	2.4	445	0.21	2.2	8.0	6.4	3–32	0.03	—
Ni	0.6	9.2	6040	1.0	0.8	450	0.55	2.4	8.0	7.9	5.8–16	0.03	0.02–0.1

TABLE I. Predicted against observed values of the linear coefficient,  $\gamma$ , in the electrical resistivity of the ferromagnetic metals at low temperatures, and the spin-lattice relaxation time  $\tau_{\downarrow\uparrow}$  at  $T = 300^\circ\text{K}$ . See Appendix A for the explanation of the appropriate values of the quantities presented here.

respectively, by estimating the spin-lattice relaxation time as  $1/\tau_{\downarrow\uparrow} = 4\pi\delta\Omega_F \text{mean}(\langle w_{\mathbf{k}\downarrow\rightarrow\mathbf{k}'\uparrow}^{\text{max}} \rangle, \langle w_{\mathbf{k}\downarrow\rightarrow\mathbf{k}'\uparrow}^{\text{min}} \rangle) = 2\pi\delta\Omega_F \langle w_{\mathbf{k}_F\uparrow\rightarrow\mathbf{k}_F\downarrow} \rangle$ , where the average  $\langle \cdot \rangle$  is taken over the solid angle between  $\mathbf{k}$  and  $\mathbf{k}'$ . The factor  $\delta\Omega_F$  is included to account for the realistic reduction ( $\delta\Omega_F < 1$ ) or increase ( $\delta\Omega_F > 1$ ) in effective solid angle from that subtended by a spherical Fermi surface ( $\delta\Omega_F = 1$ ). Since  $\langle 1/|\Delta\mathbf{k}| \rangle = 1/\text{max}(k, k')$  in (12), the factor  $\delta\Omega_F$  is understood as referring to the majority-spin Fermi surface shape. With this, the spin-lattice relaxation time is

$$\frac{1}{\tau_{\downarrow\uparrow}} = \frac{3\pi^2}{8}\delta\Omega_F \frac{\langle |A_{\mathbf{k}_F\downarrow, \mathbf{k}_F\uparrow}|^2 \rangle (\hbar\omega_c)^2}{(k_B\Theta)^4} \left(\frac{2}{z}\right)^{4/3} \frac{c_s^4}{c_L^2 c_T^2} \quad (14)$$

$$\times \frac{(k_B T)}{\hbar} \ln \left( \frac{k_B T}{\hbar c_L q_{\text{min}}} \right) + O(T^2).$$

## V. LOW-TEMPERATURE ELECTRICAL RESISTIVITY

Due to the isotropy of our model, its contribution to the ideal electrical resistivity (excluding the residual term at  $T = 0$ ) of the ferromagnetic metals at low temperatures can be calculated from the Drude-Mott formula<sup>29</sup>

$$\rho(T) = \frac{m}{e^2 z n} \frac{1}{\tau_{\downarrow\uparrow}} = \gamma T + \eta \left( \frac{T}{\Theta} \right) \ln \left( \frac{T}{\Theta} \right) + O(T^2), \quad (15)$$

with  $n = N/V$  the atomic density and the coefficients  $\gamma$  and  $\eta$  readily obtained from (14), the first term being the dominant term around liquid-helium temperatures since  $T/\Theta$  is then much smaller than  $c_s q_D / c_L q_{\text{min}}$ , with  $q_D$  the Debye cutoff, and we use a typical length  $L = 35$  nm for the largest dimension of the samples in the electrical resistivity measurements.<sup>30</sup>

In order to obtain numerical estimates, it is convenient to write  $\langle |A_{\mathbf{k}_F\uparrow, \mathbf{k}_F\downarrow}|^2 \rangle = 2\mu_B \xi H_a$ , with the Knight ratio<sup>31</sup> defined as  $\xi = \langle |\bar{\varphi}_{\mathbf{k}_F}(0)|^2 \rangle / |\psi_a(0)|^2$ , where  $\langle |\bar{\varphi}_{\mathbf{k}_F}(0)|^2 \rangle$  is the average probability density at the nucleus of electronic states on the Fermi surface, and  $\psi_a(0)$  the wavefunction at the nucleus of the outermost  $s$  electron in the free atom which, as it is known, produces a hyperfine field of magnitude  $H_a = (8\pi/3)\mu_B |\psi_a(0)|^2$ . Note that in writing  $\langle |A_{\mathbf{k}_F\uparrow, \mathbf{k}_F\downarrow}|^2 \rangle$  in this form we have assumed that  $\varphi_{\mathbf{k}_F\uparrow}(0)$  and  $\varphi_{\mathbf{k}_F\downarrow}(0)$  deviate only slightly<sup>32,33</sup> from their arithmetic mean  $\bar{\varphi}_{\mathbf{k}_F}(0)$ .

Table I summarizes the results of our theoretical predictions, which are compared with the experimental values. The observed values of the coefficient  $\gamma$  from electrical resistivity measurements are taken from the review article of Volkenshtein *et al.*<sup>2</sup> As for the spin-lattice relaxation time at  $300^\circ\text{K}$ , from the line width of the ferromagnetic resonance signal a value of about 0.1 ns is found to fit the data best<sup>34,35</sup> for Fe and Ni, although magneto-optic Kerr effect measurements<sup>36</sup> reveal a spin-lattice relaxation time between 20 ps and 40 ns for Ni, and spin-polarized photoemission experiments<sup>37</sup> reveal this to be greater than 30 ps for Fe. The author was not able to find measurements of the spin-lattice relaxation time of hcp cobalt. From our theoretical estimates, this is believed to be nearly the same as that of iron and nickel.

## VI. DISCUSSION

The agreement of our predictions with the observed values from independent experiments and for different materials indicate the existence of two crucial modes of quasiparticle motion in metals: one in which the elementary excitations of the crystal lattice transport a definite amount of spin angular momentum in the direction of propagation (like circularly polarized photons), with the displacement field of the atoms rotating perpendicular to this propagation direction; and one in which the correlated motion of the conduction electrons, brought about by the exchange interactions, ease states in which they simultaneously “belong to” different atoms for a sufficiently long time before jumping to other atoms in their band motion. In the ferromagnetic metals, the internal magnetic induction imposes a cyclotron motion on the conduction electrons, even in the absence of external magnetic fields, allowing the two kinds of quasiparticle motion to resonate at an “activation” temperature around the temperature of liquid helium.

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## Appendix A: Values of relevant physical quantities

We explain here the values of the quantities used for the predictions shown in Table I. The number of conduction electrons per atom  $z$  consistent with the observed values of the saturation magnetization, are taken from ref. [38]; the atomic densities  $n$  as well as the Fermi energies  $E_F \simeq 7$  eV for the three elements are taken from ref. [4]; the longitudinal speeds of sound  $c_L$  are taken from ref. [39], and we use  $c_T = c_L/\sqrt{2}$ , which is valid for elastically isotropic bodies (coming from the corresponding relation<sup>40</sup>  $c_{11} = 2c_{44}$  for the elastic constants, with  $c_{12} = 0$ ), for consistency with our isotropic model. The exchange splittings of the  $4s$  band  $\Delta_{\text{ex}}$  are estimated as the energy difference between the bottoms of the majority-spin and minority-spin bands at the  $\Gamma$  point (center of Brillouin zone) extracted from the band-structure calculations of ref. [41] for Fe, ref. [42] for Co and ref. [43] for Ni, with the approximate values 0.13, 0.27 and 0.05 eV, respectively; from these the temperatures  $T_{\text{res}}$  are obtained from Eq. (13). The temperatures  $T_i$  have been estimated from the well-known<sup>15</sup> magnitudes of the internal magnetic induction  $B_i$  of 22, 18, and 6 kG for Fe, Co and Ni, respectively. The low temperature limits of the Debye temperature  $\Theta$  are taken from ref. [44], and the outermost  $s$ -electron hyperfine fields in the free atom  $H_a$  from ref. [45].

The Knight ratio accounts for any deviation in hyperfine coupling from atomic behavior and may deviate from  $\xi = 1$  for two reasons:<sup>46</sup> (i) the reduction of  $s$ -character of the wavefunctions at the Fermi surface and (ii) the fact that the wavefunctions in a metal are normalized within volumes smaller than in the free atom, causing the conduction electron density in the metal greater than in the free atom. For “simple” metals the reduction tends to predominate over the normalization effect, with  $\xi$  taking values between 0.1 and 0.8.

We take both effects into account, in their simplest form, by taking  $\xi = (1/N_{3d4s})a^3/(4\pi r^3/3)$ , with  $N_{3d4s}$  the number of electrons per atom in the  $3d$  and  $4s$  subshells, with values 8, 9, and 10 for Fe, Co, and Ni, respectively,  $a$  is the lattice constant with values 2.87, 2.51 and 3.52 Å for Fe, Co and Ni, respectively,<sup>44</sup> and  $r$  is the atomic radius with values 1.26, 1.25 and 1.24 Å for Fe, Co and Ni, respectively.<sup>44</sup> Since  $|\bar{\varphi}|^2$  and  $|\psi|^2$  have units of 1/volume, the normalization effect is then taken into account by the ratio of unit cell to atomic volumes  $a^3/(4\pi r^3/3)$ , and the reduction in  $s$ -character by the factor  $1/N_{3d4s}$ , the latter because of the  $N_{3d4s}$  electrons in the  $3d$  and  $4s$  subshells of the free atom, only a fraction  $z$  remains in the  $4s$  band in the solid,<sup>38</sup> and we need to count all the electrons which can make this donation to the  $4s$  band. With this simple rule, the values of  $\xi$  shown in Table I are in the range of those for “simple” metals. Moreover, the  $4s$  contribution to the effective hyperfine fields  $\xi H_a$  obtained in this way are in agreement (except for Ni, for which it is an order of magnitude higher) with the theoretical estimates of ref. [47] aimed at explain-

ing the observed hyperfine fields in the  $3d$  ferromagnetic metals from Mössbauer and NMR experiments.

Finally, we explain our choice of factors  $\delta\Omega_F$ . For Ni (fcc structure), the sheet of the Fermi surface coming from the  $4s$  majority-spin band is, as in copper, spherical-like, with necks touching the Brillouin zone faces near the  $L$  points.<sup>48</sup> A *rough* picture of this surface may then be drawn by considering the union of a major sphere of radius  $k_{F\uparrow}$ , centered at the  $\Gamma$  point, with little spheres with such radii as to touch the major sphere and the  $L$  points, i.e. the centers of the hexagonal faces of the truncated octahedron constituting the Brillouin zone of a fcc structure. Since there are 8 such faces, we have 8 little spheres and then we need to multiply the value  $\delta\Omega_F = 1$  corresponding to the major sphere by 8, as shown in Table I. For Co (hcp structure), similar results apply due to the correspondence between energy bands (and Fermi surfaces) of the fcc and hcp structures when the hcp double zone is rotated until its  $[0001]$  axis coincides with the  $[111]$  axis of the fcc zone.<sup>48</sup> For Fe (bcc structure), the situation is much more complex since the Fermi surface corresponding to the  $4s$ -band breaks up into small regions of electron and hole pockets.<sup>48,49</sup> Nevertheless, we consider this as a reduction in effective solid angle and take for  $\delta\Omega_F$  the neutral value shown in Table I, having in mind that a more detailed investigation should not considerably change the overall result.

## Appendix B: Orbital angular momentum of atomic (electronic) system

We begin this section by discussing the orbital angular momentum operator of a conduction electron “belonging to” the  $n^{\text{th}}$  atom at a given instant of time. The displacement of this atom from the equilibrium position  $\mathbf{R}_n$  is given by

$$\begin{aligned}\hat{\mathbf{r}}_n(t) &= \sum_{\mathbf{q}\alpha} \sqrt{\frac{\hbar}{2NM\omega_{\mathbf{q}\alpha}}} \left[ \mathbf{e}_{\mathbf{q}\alpha} e^{i(\mathbf{q}\cdot\mathbf{R}_n - \omega_{\mathbf{q}\alpha}t)} \hat{a}_{\mathbf{q}\alpha} \right. \\ &\quad \left. + \mathbf{e}_{\mathbf{q}\alpha}^* e^{-i(\mathbf{q}\cdot\mathbf{R}_n - \omega_{\mathbf{q}\alpha}t)} \hat{a}_{\mathbf{q}\alpha}^\dagger \right], \\ &= \sum_{\mathbf{q}\alpha} \sqrt{\frac{\hbar}{2NM\omega_{\mathbf{q}\alpha}}} \left( e^{-i\omega_{\mathbf{q}\alpha}t} \hat{a}_{\mathbf{q}\alpha} + e^{i\omega_{\mathbf{q}\alpha}t} \hat{a}_{-\mathbf{q}\alpha}^\dagger \right) \\ &\quad \times \mathbf{e}_{\mathbf{q}\alpha} e^{i\mathbf{q}\cdot\mathbf{R}_n},\end{aligned}\tag{B1}$$

where  $M$  is the mass of the atom, and we have used  $\omega_{-\mathbf{q}\alpha} = \omega_{\mathbf{q}\alpha}$  and  $\mathbf{e}_{-\mathbf{q}\alpha}^* = \mathbf{e}_{\mathbf{q}\alpha}$  in going from the first to the second equality. The kinetic momentum of this atom is given by

$$\begin{aligned}\hat{\mathbf{p}}_n(t) &= M\dot{\hat{\mathbf{r}}}_n = -i \sum_{\mathbf{q}\alpha} \sqrt{\frac{\hbar M\omega_{\mathbf{q}\alpha}}{2N}} \left( e^{-i\omega_{\mathbf{q}\alpha}t} \hat{a}_{-\mathbf{q}\alpha} \right. \\ &\quad \left. - e^{i\omega_{\mathbf{q}\alpha}t} \hat{a}_{\mathbf{q}\alpha}^\dagger \right) \mathbf{e}_{\mathbf{q}\alpha}^* e^{-i\mathbf{q}\cdot\mathbf{R}_n}.\end{aligned}\tag{B2}$$

From these, the orbital angular momentum,  $\hat{\mathbf{L}}_n(t) = \hat{\mathbf{r}}_n(t) \times \hat{\mathbf{p}}_n(t)$ , is expressed in second quantized notation as

$$\hat{\mathbf{L}}_n(t) = \frac{\hbar}{2N} \sum_{\mathbf{q}\alpha, \mathbf{q}'\alpha'} \sqrt{\frac{\omega_{\mathbf{q}'\alpha'}}{\omega_{\mathbf{q}\alpha}}} \left( e^{-i\omega_{\mathbf{q}\alpha}t} \hat{a}_{\mathbf{q}\alpha} + e^{i\omega_{\mathbf{q}\alpha}t} \hat{a}_{-\mathbf{q}\alpha}^\dagger \right) \mathbf{S}_{\mathbf{q}\mathbf{q}'(\alpha\alpha')} \left( e^{-i\omega_{\mathbf{q}'\alpha'}t} \hat{a}_{-\mathbf{q}'\alpha'} - e^{i\omega_{\mathbf{q}'\alpha'}t} \hat{a}_{\mathbf{q}'\alpha'}^\dagger \right) e^{i(\mathbf{q}-\mathbf{q}') \cdot \mathbf{R}_n}, \quad (\text{B3})$$

where  $\mathbf{S}_{\mathbf{q}\mathbf{q}'(\alpha\alpha')} = -i(\mathbf{e}_{\mathbf{q}\alpha} \times \mathbf{e}_{\mathbf{q}'\alpha'})$ . The fact that the angular momentum of the  $n^{\text{th}}$  atom is independent of its mass implies that this angular momentum can equally well be associated to a conduction electron comoving with the atom at time  $t$ . This is the picture that we have advocated in the main article and, to the extent to which we can portrait the corresponding electronic motion, it corresponds at low temperatures to the conduction electrons being in their proper places in the atoms<sup>50</sup> for a sufficiently long time (enough for the atoms to execute part of an oscillation about their respective lattice points) until they cross the potential barrier of the crystal potential and *collectively* jump to other (then possibly unoccupied) atoms in their band motion to conduct the electricity in the presence of an electric field.

The above picture is similar to the well-known Drude picture of electronic conduction in metals but with a *finite* duration,  $\tau_c$ , of a collision of the conduction electrons with the atoms. To support this idea let us calculate the time for an atom to execute half of an oscillation about its lattice point; we estimate it as

$$\tau_c = 2R_{\text{orb}}/c_s, \quad (\text{B4})$$

with  $c_s$  being the average speed of sound, determined from  $3/c_s^3 = 2/c_T^3 + 1/c_L^3$ , and  $R_{\text{orb}}$  the root mean square value of (B1) (effective radius of atomic orbits), which in the low temperature limit becomes

$$\begin{aligned} R_{\text{orb}}^2 &= \langle |\hat{\mathbf{r}}_n(t)|^2 \rangle = \sum_{\mathbf{q}\alpha} \frac{\hbar}{2NM\omega_{\mathbf{q}\alpha}} (2\bar{N}_{\mathbf{q}\alpha} + 1) \\ &= \frac{\hbar}{2Mn} \left[ \frac{q_D^2}{4\pi^2} \left( \frac{2}{c_T} + \frac{1}{c_L} \right) + \frac{1}{2c_s^3} \left( \frac{k_B T}{\hbar} \right)^2 \right], \end{aligned} \quad (\text{B5})$$

with  $q_D$  the Debye cutoff. Around liquid-helium temperatures ( $T = 4.2^\circ\text{K}$ ) we then have  $\tau_c \sim 2$  fs for Fe, Co, and Ni (about a factor of 2 greater at  $T = 300^\circ\text{K}$ ). This is to be compared with the Drude relaxation time (average time for an electron to hit the next atom after an electron-atom collision) which is about  $10\tau_c$  or greater at low temperatures.<sup>51</sup> It is therefore very plausible for a conduction electron to spend a time of the order of  $\tau_c$  in an atom, during which its orbital angular momentum is the same as that of the atom it temporarily “belongs to”, and then jump to other atoms to conduct electricity.

We can express (B3) in the Schrödinger picture, where operators are time-independent, by means of the unitary

transformation

$$e^{i\hat{\mathcal{H}}_0 t/\hbar} a_{\mathbf{q}\alpha} e^{-i\hat{\mathcal{H}}_0 t/\hbar} = e^{-i\omega_{\mathbf{q}\alpha}t} a_{\mathbf{q}\alpha}, \quad (\text{B6})$$

$$e^{i\hat{\mathcal{H}}_0 t/\hbar} a_{\mathbf{q}\alpha}^\dagger e^{-i\hat{\mathcal{H}}_0 t/\hbar} = e^{i\omega_{\mathbf{q}\alpha}t} a_{\mathbf{q}\alpha}^\dagger, \quad (\text{B7})$$

with  $\hat{\mathcal{H}}_0$  the Hamiltonian of the uncoupled electron-phonon system. These equations can easily be derived by an iterated application of the commutation relations  $[a_{\mathbf{q}\alpha}^\dagger a_{\mathbf{q}\alpha}, a_{\mathbf{q}\alpha}] = -a_{\mathbf{q}\alpha}$  and  $[a_{\mathbf{q}\alpha}^\dagger a_{\mathbf{q}\alpha}, a_{\mathbf{q}\alpha}^\dagger] = a_{\mathbf{q}\alpha}^\dagger$ . Then  $\hat{\mathbf{L}}_n(t) = e^{i\hat{\mathcal{H}}_0 t/\hbar} (\hbar \hat{\mathbf{L}}_n) e^{-i\hat{\mathcal{H}}_0 t/\hbar}$  and the orbital angular momentum (in units of  $\hbar$ ) can be written, in the Schrödinger picture, as  $\hat{\mathbf{L}}_n = (1/N) \sum_{\mathbf{q}\mathbf{q}'} \hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}'} e^{i(\mathbf{q}-\mathbf{q}') \cdot \mathbf{R}_n}$  with

$$\begin{aligned} \hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}'} &= \frac{1}{2} \sum_{\alpha\alpha'} \sqrt{\frac{\omega_{\mathbf{q}'\alpha'}}{\omega_{\mathbf{q}\alpha}}} (\hat{a}_{\mathbf{q}\alpha} + \hat{a}_{-\mathbf{q}\alpha}^\dagger) \mathbf{S}_{\mathbf{q}\mathbf{q}'(\alpha\alpha')} \\ &\quad \times (\hat{a}_{-\mathbf{q}'\alpha'} - \hat{a}_{\mathbf{q}'\alpha'}^\dagger). \end{aligned} \quad (\text{B8})$$

Choosing  $\mathbf{e}_{\mathbf{q}1} = (1, 0, 0)$ ,  $\mathbf{e}_{\mathbf{q}2} = (0, 1, 0)$ , and  $\mathbf{e}_{\mathbf{q}3} = (0, 0, 1)$ , we can represent the matrices  $\mathbf{S}_{\mathbf{q}(\alpha\alpha')} \equiv \mathbf{S}_{\mathbf{q}\mathbf{q}(\alpha\alpha')}$  as

$$\begin{aligned} \mathbf{S}_{\mathbf{q}}^1 &= \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \quad \mathbf{S}_{\mathbf{q}}^2 = \begin{pmatrix} 0 & 0 & i \\ 0 & 0 & 0 \\ -i & 0 & 0 \end{pmatrix}, \\ \mathbf{S}_{\mathbf{q}}^3 &= \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \end{aligned} \quad (\text{B9})$$

It is convenient, however, to go to the representation where  $\mathbf{S}_{\mathbf{q}}^3$  is diagonal. This is obtained by changing basis from  $\{\mathbf{e}_{\mathbf{q}\alpha}\}$  to the helicity basis  $\{\mathbf{e}_{\mathbf{q}\sigma}\}$ . The unitary transformation performing such a change of basis is represented by

$$U_{\mathbf{q}} = \begin{pmatrix} -\frac{i}{\sqrt{2}} & 0 & \frac{i}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ 0 & i & 0 \end{pmatrix}.$$

Under this transformation, the  $\mathbf{S}_{\mathbf{q}}$  matrices transform as  $\mathbf{S}_{\mathbf{q}(\sigma\sigma')} = \sum_{\alpha\alpha'} U_{\mathbf{q}(\sigma\alpha)}^\dagger \mathbf{S}_{\mathbf{q}(\alpha\alpha')} U_{\mathbf{q}(\alpha'\sigma')}$ , which are the spin-1 matrices, with  $\bar{T}$  denoting the hermitian conjugate in spin-1 space. We have, e.g. for the third component and the ladder matrices

$$\mathcal{S}_q^3 = \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad \mathcal{S}_q^+ = \begin{pmatrix} 0 & \sqrt{2} & 0 \\ 0 & 0 & \sqrt{2} \\ 0 & 0 & 0 \end{pmatrix}, \quad \mathcal{S}_q^- = \begin{pmatrix} 0 & 0 & 0 \\ \sqrt{2} & 0 & 0 \\ 0 & \sqrt{2} & 0 \end{pmatrix}. \quad (\text{B10})$$

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With these, the angular momentum operator of a phonon  $\hat{\mathbf{S}}_{\mathbf{q},\text{ph}} = \hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}}$  can be written in matrix form, from (B8), as

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$$\begin{aligned} \hat{\mathbf{S}}_{\mathbf{q},\text{ph}} &= \frac{1}{2} \begin{pmatrix} \frac{1}{\sqrt{\omega_{q1}}} (\hat{a}_{q1} + \hat{a}_{-q1}^\dagger) \\ \frac{1}{\sqrt{\omega_{q2}}} (\hat{a}_{q2} + \hat{a}_{-q2}^\dagger) \\ \frac{1}{\sqrt{\omega_{q3}}} (\hat{a}_{q3} + \hat{a}_{-q3}^\dagger) \end{pmatrix}^{\bar{T}} \mathcal{S}_q \begin{pmatrix} \sqrt{\omega_{q1}} (\hat{a}_{-q1} - \hat{a}_{q1}^\dagger) \\ \sqrt{\omega_{q2}} (\hat{a}_{-q2} - \hat{a}_{q2}^\dagger) \\ \sqrt{\omega_{q3}} (\hat{a}_{-q3} - \hat{a}_{q3}^\dagger) \end{pmatrix} \\ &= \frac{1}{2} \begin{pmatrix} \frac{1}{\sqrt{\omega_{q1}}} (\hat{a}_{q1} + \hat{a}_{-q1}^\dagger) \\ \frac{1}{\sqrt{\omega_{q2}}} (\hat{a}_{q2} + \hat{a}_{-q2}^\dagger) \\ \frac{1}{\sqrt{\omega_{q3}}} (\hat{a}_{q3} + \hat{a}_{-q3}^\dagger) \end{pmatrix}^{\bar{T}} U_q \mathcal{S}_q U_q^{\bar{T}} \begin{pmatrix} \sqrt{\omega_{q1}} (\hat{a}_{-q1} - \hat{a}_{q1}^\dagger) \\ \sqrt{\omega_{q2}} (\hat{a}_{-q2} - \hat{a}_{q2}^\dagger) \\ \sqrt{\omega_{q3}} (\hat{a}_{-q3} - \hat{a}_{q3}^\dagger) \end{pmatrix} \\ &= \frac{1}{2} \begin{pmatrix} \frac{1}{\sqrt{\omega_{qT}}} (\hat{b}_{q-} + \hat{b}_{-q-}^\dagger) \\ -\frac{1}{\sqrt{\omega_{qL}}} (\hat{b}_{q0} + \hat{b}_{-q0}^\dagger) \\ \frac{1}{\sqrt{\omega_{qT}}} (\hat{b}_{q+} + \hat{b}_{-q+}^\dagger) \end{pmatrix}^{\bar{T}} \mathcal{S}_q \begin{pmatrix} \sqrt{\omega_{qT}} (\hat{b}_{-q-} - \hat{b}_{q-}^\dagger) \\ -\sqrt{\omega_{qL}} (\hat{b}_{-q0} - \hat{b}_{q0}^\dagger) \\ \sqrt{\omega_{qT}} (\hat{b}_{-q+} - \hat{b}_{q+}^\dagger) \end{pmatrix}, \end{aligned} \quad (\text{B11})$$

where we have introduced the operators

$$\hat{b}_{q\pm} = \frac{1}{\sqrt{2}} (\hat{a}_{q2} \mp i \hat{a}_{q1}), \quad \text{and} \quad \hat{b}_{q0} = i \hat{a}_{q3}, \quad (\text{B12})$$

which satisfy the bosonic commutation relations. Using (B10) we have

$$\begin{aligned} \hat{S}_{\mathbf{q},\text{ph}}^- &= \frac{1}{2} \left[ \sqrt{\frac{2\omega_{qL}}{\omega_{qT}}} (\hat{b}_{q-}^\dagger + \hat{b}_{q+}^\dagger) (\hat{b}_{q0}^\dagger + \hat{b}_{q0}) \right. \\ &\quad \left. - \sqrt{\frac{2\omega_{qT}}{\omega_{qL}}} (\hat{b}_{q0}^\dagger - \hat{b}_{q0}) (\hat{b}_{q-}^\dagger - \hat{b}_{q+}^\dagger) \right], \end{aligned} \quad (\text{B13})$$

$$\hat{S}_{\mathbf{q},\text{ph}}^3 = \hat{b}_{q+}^\dagger \hat{b}_{q+} - \hat{b}_{q-}^\dagger \hat{b}_{q-}, \quad (\text{B14})$$

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with  $\hat{S}_{\mathbf{q},\text{ph}}^+ = (\hat{S}_{\mathbf{q},\text{ph}}^-)^\dagger$ . Here, we have used the relations (inherited from  $\epsilon_{-q\sigma}^* = \epsilon_{q\sigma}$ )

$$\hat{b}_{-q\sigma}^* = \hat{b}_{q\sigma} \Leftrightarrow \hat{b}_{-q\sigma} = \hat{b}_{q\sigma}^* = \hat{b}_{q\bar{\sigma}}. \quad (\text{B15})$$

Note that, in spin space, the  $\hat{a}_{q\alpha}$  are treated just as real numbers and then the  $\hat{b}_{q\sigma}$  as complex numbers (in both cases conserving the order of multiplication in the matrix products), with  $\bar{\sigma} = \{-, +\}$  according to  $\sigma = \{+, -\}$ , respectively, and  $\hat{b}_{q0} = -\hat{b}_{q0}$ . Eq. (B15) can also be written as  $\mathcal{PT} \hat{b}_{q\sigma} = \hat{b}_{q\sigma}$ , where  $\mathcal{P}$  is the parity operator and  $\mathcal{T}$  the complex conjugation. This states that all the wavefunctions of the crystal vibrations are even under the  $\mathcal{PT}$  transformation.

We can follow the same procedure leading to (B11) with  $\hat{\mathbf{S}}_{\mathbf{q}\mathbf{q}'}$  for  $\mathbf{q} \neq \mathbf{q}'$ . In this case, we write (B8) as



$$\begin{aligned}
\hat{S}_{qq'} &= \frac{1}{2} \begin{pmatrix} \frac{1}{\sqrt{\omega_{q1}}} (\hat{a}_{q1} + \hat{a}_{-q1}^\dagger) \\ \frac{1}{\sqrt{\omega_{q2}}} (\hat{a}_{q2} + \hat{a}_{-q2}^\dagger) \\ \frac{1}{\sqrt{\omega_{q3}}} (\hat{a}_{q3} + \hat{a}_{-q3}^\dagger) \end{pmatrix}^{\bar{T}} S_{qq'} \begin{pmatrix} \sqrt{\omega_{q'1}} (\hat{a}_{-q'1} - \hat{a}_{q'1}^\dagger) \\ \sqrt{\omega_{q'2}} (\hat{a}_{-q'2} - \hat{a}_{q'2}^\dagger) \\ \sqrt{\omega_{q'3}} (\hat{a}_{-q'3} - \hat{a}_{q'3}^\dagger) \end{pmatrix}, \\
&= \frac{1}{2} \begin{pmatrix} \frac{1}{\sqrt{\omega_{qT}}} (\hat{b}_{q-} + \hat{b}_{-q+}^\dagger) \\ \frac{1}{\sqrt{\omega_{qL}}} (\hat{b}_{-q0}^\dagger - \hat{b}_{q0}) \\ \frac{1}{\sqrt{\omega_{qT}}} (\hat{b}_{q+} + \hat{b}_{-q-}^\dagger) \end{pmatrix}^{\bar{T}} \tilde{U}_{qq'}^{\bar{T}} S_{qq'} \tilde{U}_{q'q} \begin{pmatrix} \sqrt{\omega_{q'T}} (\hat{b}_{-q'-} - \hat{b}_{q'-}^\dagger) \\ \sqrt{\omega_{q'L}} (\hat{b}_{q'0}^\dagger - \hat{b}_{-q'0}) \\ \sqrt{\omega_{q'T}} (\hat{b}_{-q'+} - \hat{b}_{q'+}^\dagger) \end{pmatrix}
\end{aligned} \tag{B16}$$

where we have defined  $\tilde{U}_{qq'} = U_q \otimes 1_{q'}$  and  $\tilde{U}_{q'q} = 1_q \otimes U_{q'}$  in the tensor product spin space corresponding to the phonons with  $q$  and  $q'$ . Since  $\tilde{U}_{qq'}^{\bar{T}} S_{qq'} \tilde{U}_{q'q}$  must transform as an angular momentum matrix upon rotations and must equal  $S_q$  for  $q' = q$  we take  $\tilde{U}_{qq'}^{\bar{T}} S_{qq'} \tilde{U}_{q'q} = S_q \otimes 1_{q'} = 1_q \otimes S_{q'}$ . With this we have, similar to (B13)

$$\begin{aligned}
\hat{S}_{qq'}^- &= \frac{1}{2} \left[ \sqrt{\frac{2\omega_{q'L}}{\omega_{qT}}} (\hat{b}_{q-}^\dagger + \hat{b}_{q+}) (\hat{b}_{q'0}^\dagger + \hat{b}_{q'0}) \right. \\
&\quad \left. - \sqrt{\frac{2\omega_{q'T}}{\omega_{qL}}} (\hat{b}_{q0}^\dagger - \hat{b}_{q0}) (\hat{b}_{q'-}^\dagger - \hat{b}_{q'+}) \right],
\end{aligned} \tag{B17}$$

which is Eq. (7) in the main article.

## Appendix C: Transition probability rates and spin-lattice relaxation time

### 1. Temperature-dependent part

The perturbation mediating the spin-flip transitions restoring the initial equilibrium imposed by the exchange

interactions is mainly

$$\hat{H}_c^{\uparrow\downarrow} = \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}', qq'} A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow} \delta_{\Delta\mathbf{q}, -\Delta\mathbf{k}} \hat{c}_{\mathbf{k}'\uparrow}^\dagger \hat{c}_{\mathbf{k}\downarrow} \hat{S}_{qq'}^-, \tag{C1}$$

with  $\hat{S}_{qq'}^-$  given by (B17). We first consider processes conserving the total number of phonons. Those which do not conserve it either are not allowed by conservation of energy (as is the case for the processes where a pair of phonons are absorbed, coming from the terms containing  $\hat{b}_{q+}\hat{b}_{q'0}$  and  $\hat{b}_{q0}\hat{b}_{q'+}$  in (B17)) or give exponentially small contributions at low temperatures (as is the case for the processes where a pair of phonons are emitted, coming from the terms  $\hat{b}_{q-}^\dagger\hat{b}_{q'0}^\dagger$  and  $\hat{b}_{q0}^\dagger\hat{b}_{q'-}^\dagger$  in (B17)), except for the temperature-independent part arising from the *spontaneous* emission of a pair of phonons in the electronic transitions, which we treat in the next section.

Since the thermal average number of phonons in a given state decreases exponentially at very low temperature except for the state with minimum energy, the most probable transitions are those in which one of the phonons, in the processes conserving the total number, is emitted *spontaneously*. The rate of these transitions, per conduction electron, from the occupied state  $\mathbf{k}\downarrow$  to the unoccupied state  $\mathbf{k}'\uparrow$  is, in the leading order of perturbation theory,

$$\begin{aligned}
w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow} &= \frac{2\pi}{\hbar} \frac{1}{4^2} |A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2 \frac{1}{N_e} \sum_q \left\{ \frac{2\omega_{qL}}{\omega_{|\Delta\mathbf{k}-\mathbf{q}|T}} \left[ \bar{N}_{q0} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - \hbar c_L q + \Delta\varepsilon_{\mathbf{k}}) \right. \right. \\
&\quad \left. \left. + \bar{N}_{|\Delta\mathbf{k}-\mathbf{q}|+} \delta(\hbar c_L q - \hbar c_T |\Delta\mathbf{k} - \mathbf{q}| + \Delta\varepsilon_{\mathbf{k}}) \right] + \frac{2\omega_{|\Delta\mathbf{k}-\mathbf{q}|T}}{\omega_{qL}} \left[ \bar{N}_{|\Delta\mathbf{k}-\mathbf{q}|+} \delta(\hbar c_L q - \hbar c_T |\Delta\mathbf{k} - \mathbf{q}| + \Delta\varepsilon_{\mathbf{k}}) \right. \right. \\
&\quad \left. \left. + \bar{N}_{q0} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - \hbar c_L q + \Delta\varepsilon_{\mathbf{k}}) \right] \right\},
\end{aligned} \tag{C2}$$

where  $\Delta\varepsilon_{\mathbf{k}} = \varepsilon_{\mathbf{k}'} - \varepsilon_{\mathbf{k}} < 0$  (spin- $\downarrow$  electrons loose energy in the process of restoring the initial equilib-

rium imposed by the exchange interactions),  $\bar{N}_{q\sigma}$  is the Bose distribution,  $N_e$  is the total number of conduction

electrons, and  $q = |\mathbf{q}|$ . In the thermodynamic limit,  $\sum_{\mathbf{q}} \rightarrow (V/8\pi^3) \int d^3\mathbf{q}$ , with  $V$  the volume of the ferromagnetic body. For the integration, we use spherical coordinates with  $\Delta\mathbf{k}$  in the direction of the 3-axis. Denoting  $x = \cos(\Delta\mathbf{k}, \mathbf{q})$ , we can write the  $\delta$ -functions, for fixed values of  $\Delta\mathbf{k}$  and  $\Delta\varepsilon_{\mathbf{k}}$ , as

$$\delta(f_q(x)) = \frac{\delta(x - \bar{x}_{q_r})}{|df_q(x)/dx|_{\bar{x}_{q_r}}}, \quad (C3)$$

$$f_q(x) = s_T \hbar c_T |\Delta\mathbf{k} - \mathbf{q}|_x + s_L \hbar c_L q + \Delta\varepsilon_{\mathbf{k}},$$

where  $s_L$  and  $s_T$  (with  $s_L = -s_T$ ) are the signs appearing

in the arguments of the  $\delta$ -functions in (12), and  $\bar{x}_{q_r}$  is the value of  $x$  for which conservation of energy  $f_q(\bar{x}_{q_r}) = 0$  holds, given the value of  $q = q_r$ . From (C3) we have

$$|df_q(x)/dx|_x^{-1} = \frac{|\Delta\mathbf{k} - \mathbf{q}|_x}{\hbar c_T |\Delta\mathbf{k}| q}, \quad (C4)$$

$$|\Delta\mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}} = \left| \frac{c_L}{c_T} q_r - \frac{\Delta\varepsilon_{\mathbf{k}}}{\hbar c_T} \right|.$$

Therefore we can write (C2), in the thermodynamic limit, as

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$$w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow} = \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2}{16\pi\hbar} \frac{V}{N_e} \frac{1}{\hbar c_T |\Delta\mathbf{k}|} \int_{q_{\min}}^{q_{\max}} q^2 dq \int_{-1}^1 dx \left\{ \frac{c_L q}{c_T |\Delta\mathbf{k} - \mathbf{q}|_x} \left[ \bar{N}_{q0} \delta(x - \bar{x}_{q_r}) \frac{|\Delta\mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right. \right. \\ \left. \left. + \bar{N}_{|\Delta\mathbf{k} - \mathbf{q}|_x} \delta(x - \bar{x}_{q_r}) \frac{|\Delta\mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right] + \frac{c_T |\Delta\mathbf{k} - \mathbf{q}|_x}{c_L q} \left[ \bar{N}_{|\Delta\mathbf{k} - \mathbf{q}|_x} \delta(x - \bar{x}_{q_r}) \frac{|\Delta\mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right. \right. \\ \left. \left. + \bar{N}_{q0} \delta(x - \bar{x}_{q_r}) \frac{|\Delta\mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right] \right\}. \quad (C5)$$


---

We are now allowed to collapse the  $\delta$ -functions provided  $\bar{x}_{q_r} \in [-1, 1]$  exists, for at least one value of  $q = q_r$  in the range  $q \in [q_{\min}, q_{\max}]$ , given the quantities  $\Delta\mathbf{k}$  and  $\Delta\varepsilon_{\mathbf{k}}$ , that is, if conservation of energy  $f_q(\bar{x}_{q_r}) = 0$  applies. We can classify the terms in (C2) as those for which  $s_T = 1$  and those for which  $s_T = -1$ . Since  $\Delta\varepsilon_{\mathbf{k}} < 0$ , the family of energy-conserving surfaces in  $q$ -space (concentric spheres) lie, for the first class ( $s_T = 1$ ),

at and continuously between

$$\hbar c_T |\Delta\mathbf{k}| - \hbar(c_L - c_T)q_r - |\Delta\varepsilon_{\mathbf{k}}| = 0, \quad (C6)$$

$$\hbar c_T |\Delta\mathbf{k}| - \hbar(c_L + c_T)q_r - |\Delta\varepsilon_{\mathbf{k}}| = 0, \quad (C7)$$

these two being the ones for which  $\bar{x}_{q_r} = -1$  and  $\bar{x}_{q_r} = 1$ , respectively. The general equation for these spheres is

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$$q_r = \frac{\bar{x}_{q_r} |\Delta\mathbf{k}| + (c_L/c_T)(|\Delta\varepsilon_{\mathbf{k}}|/\hbar c_T)}{(c_L/c_T)^2 - 1} \left[ \pm \sqrt{1 - \frac{[(c_L/c_T)^2 - 1][(|\Delta\varepsilon_{\mathbf{k}}|/\hbar c_T)^2 - |\Delta\mathbf{k}|^2]}{[\bar{x}_{q_r} |\Delta\mathbf{k}| + (c_L/c_T)(|\Delta\varepsilon_{\mathbf{k}}|/\hbar c_T)]^2}} - 1 \right], \quad (C8)$$


---

where the  $+$  or  $-$  in front of the square root is chosen such that  $q_r$  is always positive for every value of  $\bar{x}_{q_r} \in [-1, 1]$ . It is then seen that  $q_r$  increases monotonically with  $\bar{x}_{q_r}$  from a value  $q_r = q_{\min} = 0^+$  (for  $\bar{x}_{q_r} = 1$ ) to a value  $q_r = q_{\max}$  (for  $\bar{x}_{q_r} = -1$ ) given, from (C6), by

$$q_{\max} = \frac{2c_T}{c_L - c_T} k_F = (z/2)^{1/3} \frac{2c_T}{c_L - c_T} q_D, \quad (C9)$$

with  $z = N_e/N$  the number of conduction electrons per atom. Since  $0 \leq |\Delta\varepsilon_{\mathbf{k}}| \leq \hbar\omega_c$  and  $\Delta k_F \leq |\Delta\mathbf{k}| \leq 2k_F$ , the lower limit for  $q$  corresponds, from (C6) and (C7), to

$$\hbar c_T |\Delta\mathbf{k}| - |\Delta\varepsilon_{\mathbf{k}}| = 0, \quad (C10)$$

up to a negligible term of order  $O(1/L)$ ; which is satisfied when  $|\Delta\varepsilon_{\mathbf{k}}| = |\Delta\varepsilon_{\mathbf{k}}|_{\max} = \hbar\omega_c$  and  $|\Delta\mathbf{k}| = |\Delta\mathbf{k}|_{\min} = \Delta k_F$ , as discussed in the main article. The upper limit is realized when  $|\Delta\varepsilon_{\mathbf{k}}| = |\Delta\varepsilon_{\mathbf{k}}|_{\min} = 0$  and  $|\Delta\mathbf{k}| = |\Delta\mathbf{k}|_{\max} = 2k_F$  corresponding to electronic backscattering at the Fermi surface. Therefore, for the processes in (C2) for which  $s_T = 1$ , conservation of energy applies for each value of  $q \in [q_{\min}, q_{\max}]$ , that is,  $\bar{x}_{q_r}$  can always be found by inverting (C8). This indicates that the integration of the first and last terms in (C5) can be done by first integrating over  $x$  (collapsing the  $\delta$ -functions) and then integrating the result over  $q$ .

For the second class of processes ( $s_T = -1$ ), the family

of energy-conserving surfaces in  $q$ -space lie at and continuously between

$$\hbar(c_L + c_T)q_r - \hbar c_T|\Delta\mathbf{k}| - |\Delta\varepsilon_{\mathbf{k}}| = 0, \quad (\text{C11})$$

$$\hbar(c_L - c_T)q_r - \hbar c_T|\Delta\mathbf{k}| - |\Delta\varepsilon_{\mathbf{k}}| = 0. \quad (\text{C12})$$

However, in these cases, it is not allowed by conservation

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$$\begin{aligned} w_{\mathbf{k}\downarrow\rightarrow\mathbf{k}'\uparrow} &= \frac{|A_{\mathbf{k}\downarrow,\mathbf{k}'\uparrow}|^2}{16\pi\hbar|\Delta\mathbf{k}|} \frac{V}{N_e} \frac{c_L}{c_T} \frac{1}{\hbar c_T} \left[ \int_{q_{\min}}^{q_{\max}} dq \frac{q^2}{e^{\beta\hbar c_L q} - 1} + (c_T/c_L)^2 \int_{q_{\min}}^{q_{\max}} dq \frac{|(c_L/c_T)q + |\Delta\varepsilon_{\mathbf{k}}|/\hbar c_T|^2}{e^{\beta\hbar c_L q} - 1} \right], \\ &= \frac{|A_{\mathbf{k}\downarrow,\mathbf{k}'\uparrow}|^2}{16\pi\hbar|\Delta\mathbf{k}|} \frac{V}{N_e} \frac{c_L}{c_T} \frac{1}{\hbar c_T} \frac{1}{(\hbar c_L \beta)^3} \left[ \int_{\beta\hbar c_L q_{\min}}^{\beta\hbar c_L q_{\max}} dy \frac{y^2}{e^y - 1} + \int_{\beta\hbar c_L q_{\min}}^{\beta\hbar c_L q_{\max}} dy \frac{(y + \beta|\Delta\varepsilon_{\mathbf{k}}|)^2}{e^y - 1} \right], \end{aligned} \quad (\text{C13})$$


---

where  $\beta^{-1} = k_B T$ . Since  $\hbar c_s q_D = k_B \Theta$  and  $q_D = (6\pi^2 n)^{1/3} = (2/z)^{1/3} k_F$ , we can write

$$\frac{1}{c_T^2 c_L^2 \hbar^4} = \frac{c_s^4}{c_T^2 c_L^2} \frac{6\pi^2 n (2/z)^{1/3} k_F}{(k_B \Theta)^4}, \quad (\text{C14})$$

with  $n = N/V$  the atomic density. Therefore, for  $T \rightarrow 0$ , we have for the temperature-dependent transition probability rate, per conduction electron, of scattering from the occupied state  $\mathbf{k}\downarrow$  to the unoccupied state  $\mathbf{k}'\uparrow$

$$\begin{aligned} w_{\mathbf{k}\downarrow\rightarrow\mathbf{k}'\uparrow} &= \frac{3\pi}{16} \frac{|A_{\mathbf{k}\downarrow,\mathbf{k}'\uparrow}|^2 |\Delta\varepsilon_{\mathbf{k}}|^2}{(k_B \Theta)^4} \frac{k_F}{|\Delta\mathbf{k}|} \frac{c_s^4}{c_L^2 c_T^2} \left( \frac{2}{z} \right)^{4/3} \\ &\times \frac{(k_B T)}{\hbar} \ln \left( \frac{k_B T}{\hbar c_L q_{\min}} \right) + O(T^2), \end{aligned} \quad (\text{C15})$$

which is Eq. (12) in the main article, where we have used the fact that  $\int_a^b dy/(e^y - 1) \rightarrow \ln(1/a)$  when  $b \rightarrow \infty$ .

Having sketched the calculation of the transition probability rate for the processes  $\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow$ , we now dis-

cus the decay time of a spin- $\downarrow$  electron in state  $\mathbf{k}$ , with  $E_{F\uparrow} \leq \varepsilon_{\mathbf{k}} \leq E_{F\downarrow}$ . In the leading order of perturbation theory, this is

$$\frac{1}{\tau_{\mathbf{k}\downarrow}} = \sum_{\mathbf{k}' (k' = k_{F\uparrow})} [f_{\mathbf{k}'\uparrow}^0 w_{\mathbf{k}'\uparrow\rightarrow\mathbf{k}\downarrow} + w_{\mathbf{k}\downarrow\rightarrow\mathbf{k}'\uparrow} (1 - f_{\mathbf{k}'\uparrow}^0)], \quad (\text{C16})$$

where  $f_{\mathbf{k}s}^0 = \{\exp[(\varepsilon_{\mathbf{k}} - \mu_s(T))/k_B T] + 1\}^{-1}$  is the Fermi-Dirac (local) equilibrium distribution for the electrons of spin  $s$ , with  $\mu_s(T)$  being the corresponding chemical potential, with  $\mu_s(0) = E_{Fs}$ . Note that  $E_{F\uparrow} \neq E_{F\downarrow}$  due to the initial Fermi surface instability. Since we are interested only in the leading contribution to the electrical resistivity at low temperatures we extract from (C16) the dominant term, which has the desired temperature dependence. First, the transition probability rate of the (inverse) processes  $\mathbf{k}\uparrow \rightarrow \mathbf{k}'\downarrow$  (in which the electrons gain energy) can be written, similar to (C2), as

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$$\begin{aligned} w_{\mathbf{k}\uparrow\rightarrow\mathbf{k}'\downarrow} &= \frac{2\pi}{\hbar} \frac{1}{4^2} |A_{\mathbf{k}\uparrow,\mathbf{k}'\downarrow}|^2 \frac{1}{N_e} \sum_{\mathbf{q}} \left\{ \frac{2\omega_{qL}}{\omega_{|\Delta\mathbf{k}-\mathbf{q}|T}} [\bar{N}_{q0} \delta(\hbar c_L q - \hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - |\Delta\varepsilon_{\mathbf{k}}|) \right. \\ &\quad + \bar{N}_{|\Delta\mathbf{k}-\mathbf{q}|} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - \hbar c_L q - |\Delta\varepsilon_{\mathbf{k}}|)] + \frac{2\omega_{|\Delta\mathbf{k}-\mathbf{q}|T}}{\omega_{qL}} [\bar{N}_{|\Delta\mathbf{k}-\mathbf{q}|} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - \hbar c_L q - |\Delta\varepsilon_{\mathbf{k}}|) \\ &\quad \left. + \bar{N}_{q0} \delta(\hbar c_L q - \hbar c_T |\Delta\mathbf{k} - \mathbf{q}| - |\Delta\varepsilon_{\mathbf{k}}|)] \right\}. \end{aligned} \quad (\text{C17})$$


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However, it is observed now that the  $s_T = 1$  processes give exponentially small contributions at low temperatures and then these transitions are unlikely, so we ignore them from (C16). The resulting quasi-unidirectional

transitions just corroborate the insight of spin- $\downarrow$  electrons “decaying” to the majority-spin band to restore the initial equilibrium imposed by the exchange interactions (see Fig. 3). The remaining sum in (C16) may be split

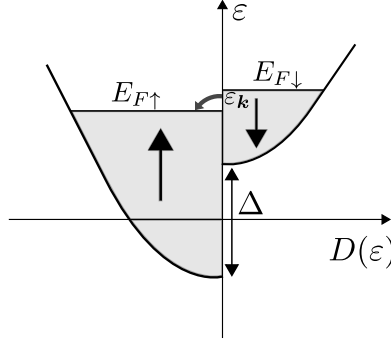


FIG. 3. Sketch of the initial instability caused by coupling a magnetic field to electrons which have reached equilibrium under exchange interactions (in ferromagnets, this field is internal to the materials). To restore this equilibrium at low temperatures, a typical spin- $\downarrow$  electron in state  $\mathbf{k}$  with energy  $\varepsilon_{\mathbf{k}} > E_{F\uparrow}$  will most likely decay to unoccupied states with energy  $E_{F\uparrow}$ .

as  $\sum_{\mathbf{k}'} w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow} - \sum_{\mathbf{k}'} f_{\mathbf{k}'\uparrow}^0 w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}$ . From this, the first sum dominates since  $f_{\mathbf{k}'\uparrow}^0 \in [0, 1]$ . The second sum has the temperature dependence of the first sum but multiplied by additional factors of temperature coming from  $f_{\mathbf{k}'\uparrow}^0$ ; we write the relatively small contributions coming from this with ellipsis when calculating the *average* relaxation time of a spin- $\downarrow$  electron. This is defined as

$$\begin{aligned} \frac{1}{\tau_{\downarrow\uparrow}} &= \left\langle \frac{1}{\tau_{\mathbf{k}\downarrow}} \right\rangle_{0 \leq \varepsilon_{\mathbf{k}} \leq \hbar\omega_c} \\ &= \sum_{\mathbf{k}' (k'=k_{F\uparrow})} \langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow} \rangle_{0 \leq \varepsilon_{\mathbf{k}} \leq \hbar\omega_c} + \dots \\ &= 4\pi\delta\Omega_F \text{mean}(\langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\max} \rangle, \langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\min} \rangle) + \dots \end{aligned} \quad (\text{C18})$$

Here  $\langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\max} \rangle$  is the average of (C15) over the solid angle made by  $\mathbf{k}$  and  $\mathbf{k}'$ , evaluated at  $|\Delta\varepsilon_{\mathbf{k}}| = |\Delta\varepsilon_{\mathbf{k}}|_{\max} = \hbar\omega_c$  and  $|\Delta\mathbf{k}| = |\Delta\mathbf{k}|_{\min} = \Delta k_F$ , and  $\langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\min} \rangle$  is the same average evaluated at  $|\Delta\varepsilon_{\mathbf{k}}| = |\Delta\varepsilon_{\mathbf{k}}|_{\min} = 0$  and  $|\Delta\mathbf{k}| = |\Delta\mathbf{k}|_{\max} = 2k_F$ . Note that  $\int d\Omega_{\mathbf{k}, \mathbf{k}'} / |\Delta\mathbf{k}| = 4\pi / \max(k, k')$  and then  $\langle k_F / |\Delta\mathbf{k}|_{\min/\max} \rangle = k_F / k_{F\uparrow} = 1 + O(\Delta_{\text{ex}}/2E_F)$ , which is negligibly deviating from unity. Also, we have introduced the factor  $\delta\Omega_F$  accounting for the reduction or increase in solid angle due to the realistic deviation of the shape of the majority-spin fermi surface

from a sphere (for which  $\delta\Omega_F = 1$ ). With this, (C18) becomes

$$\begin{aligned} \frac{1}{\tau_{\downarrow\uparrow}} &= \frac{3\pi^2}{8} \delta\Omega_F \frac{\langle |A_{k_{F\downarrow}, k_{F\uparrow}}|^2 \rangle (\hbar\omega_c)^2}{(k_B\Theta)^4} \left(\frac{2}{z}\right)^{4/3} \frac{c_s^4}{c_L^2 c_T^2} \\ &\quad \times \frac{(k_B T)}{\hbar} \ln \left( \frac{k_B T}{\hbar c_L q_{\min}} \right) + O(T^2), \end{aligned} \quad (\text{C19})$$

in which the ellipsis in (C18) are absorbed in the notation  $O(T^2)$ . This is Eq. (14) in the main article, from which the electrical resistivity readily follows. Note that in obtaining the linear in temperature contribution to the resistivity we use in (C15)

$$\ln \left( \frac{k_B T}{\hbar c_L q_{\min}} \right) = \ln \left( \frac{c_s q_D}{c_L q_{\min}} \right) + \ln \left( \frac{T}{\Theta} \right), \quad (\text{C20})$$

the first term being much greater than the second for the temperatures of interest and the typical largest length scale  $L = 35$  nm of samples in experiments.

## 2. Temperature-independent part

For the processes where a pair of phonons are *spontaneously* emitted, the transition probability rate is

$$\begin{aligned} w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\text{sp-sp}} &= \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2}{16\pi\hbar} \frac{V}{N_e} \int_{q_{\min}}^{q_{\max}} q^2 dq \int_{-1}^1 dx \left[ \frac{c_L q}{c_T |\Delta\mathbf{k} - \mathbf{q}|_x} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}|_x + \hbar c_L q + \Delta\varepsilon_{\mathbf{k}}) \right. \\ &\quad \left. + \frac{c_T |\Delta\mathbf{k} - \mathbf{q}|_x}{c_L q} \delta(\hbar c_T |\Delta\mathbf{k} - \mathbf{q}|_x + \hbar c_L q + \Delta\varepsilon_{\mathbf{k}}) \right] \end{aligned} \quad (\text{C21})$$

The family of energy-conserving surfaces in  $q$ -space lie, in these cases, at and continuously between

$$\hbar c_T |\Delta\mathbf{k}| + \hbar(c_L - c_T)q_r - |\Delta\varepsilon_{\mathbf{k}}| = 0 \quad (\text{C22})$$

$$\hbar c_T |\Delta\mathbf{k}| + \hbar(c_L + c_T)q_r - |\Delta\varepsilon_{\mathbf{k}}| = 0. \quad (\text{C23})$$

Since  $\hbar c_T |\Delta\mathbf{k}|_{\min}$  match  $|\Delta\varepsilon_{\mathbf{k}}|_{\max}$ , it is easily seen that any “finite” value of  $q$  violates (C22) and (C23). This means that in collapsing the  $\delta$ -functions in (C21) the value  $q = q_{\min}$  is selected in the integrand. That is,



with

$$|\Delta \mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}} = \left| \frac{c_L}{c_T} q_r + \frac{\Delta \varepsilon_{\mathbf{k}}}{\hbar c_T} \right|, \quad (\text{C24})$$

we have, neglecting terms proportional to powers of  $q_{\min}$ ,

$$\begin{aligned} w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\text{sp-sp}} &= \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2}{16\pi\hbar} \frac{V}{N_e} \frac{1}{\hbar c_T |\Delta \mathbf{k}|} \int_{q_{\min}}^{q_{\max}} q^2 dq \int_{-1}^1 dx \left[ \frac{c_L q}{c_T |\Delta \mathbf{k} - \mathbf{q}|_x} \delta(x - \bar{x}_{q_r}) \frac{|\Delta \mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right. \\ &\quad \left. + \frac{c_T |\Delta \mathbf{k} - \mathbf{q}|_x}{c_L q} \delta(x - \bar{x}_{q_r}) \frac{|\Delta \mathbf{k} - \mathbf{q}|_{\bar{x}_{q_r}}}{q_r} \right], \quad (\text{C25}) \\ &= \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2}{16\pi\hbar} \frac{V}{N_e} \frac{1}{\hbar c_T |\Delta \mathbf{k}|} \frac{c_L}{c_T} \left( \frac{|\Delta \varepsilon_{\mathbf{k}}|}{\hbar c_L} \right)^3 = \frac{3\pi}{16} \frac{|A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2 |\Delta \varepsilon_{\mathbf{k}}|^3}{\hbar (k_B \Theta)^4} \frac{k_F}{|\Delta \mathbf{k}|} \frac{c_s^4}{c_T^2 c_L^2} \left( \frac{2}{z} \right)^{4/3}, \end{aligned}$$

where (C14) was used. We can then define  $1/\tau_{\downarrow\uparrow}^0 = 4\pi\delta\Omega_F \text{mean}(\langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\text{sp-sp (max)}} \rangle, \langle w_{\mathbf{k}\downarrow \rightarrow \mathbf{k}'\uparrow}^{\text{sp-sp (min)}} \rangle)$  and get

$$\frac{1}{\tau_{\downarrow\uparrow}^0} = \frac{3\pi^2}{8} \delta\Omega_F \frac{\langle |A_{\mathbf{k}\downarrow, \mathbf{k}'\uparrow}|^2 \rangle (\hbar\omega_c)^3}{\hbar (k_B \Theta)^4} \frac{c_s^4}{c_T^2 c_L^2} \left( \frac{2}{z} \right)^{4/3}, \quad (\text{C26})$$

for the inverse of the leading temperature-independent contribution  $\tau_{\downarrow\uparrow}^0$  to the spin-lattice relaxation time. This is of the order of  $10^{-7}$ s for Fe, Co and Ni and then the total leading contribution  $\tau_{\downarrow\uparrow}^{\text{tot}}$  to the spin-lattice relaxation time, defined as  $1/\tau_{\downarrow\uparrow}^{\text{tot}} = 1/\tau_{\downarrow\uparrow} + 1/\tau_{\downarrow\uparrow}^0$ , is dominated around liquid-helium temperatures by the temperature-

dependent part, as shown in the main article, since  $\tau_{\downarrow\uparrow}^0/\tau_{\downarrow\uparrow} \sim 10^4$ .

It is well-known that for the ferromagnetic metals the residual resistivity is not only due to impurities and other lattice defects but that an anomalous contribution from the internal magnetic induction is also present,<sup>2,15</sup> apart from magnetostriction and magnetocrystal contributions. There has been no theory so far describing the contribution from the internal magnetic induction. From (C26) this is given by

$$\rho_0 = \frac{m}{e^2 z n} \frac{1}{\tau_{\downarrow\uparrow}^0}, \quad (\text{C27})$$

which is of the order of  $10^{-12} \Omega \text{ cm}$  for Fe, Co and Ni.

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